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IMPROVING THE REPRODUCIBILITY OF THE RADIAL ARGON CONCENTRATION IN BERYLLIUM SHELLS

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Beryllium is one of the preferred ablator materials for achieving ignition at the National Ignition Facility (NIF). Sputter coating of beryllium on spherical mandrels has been used at LLNL and at General Atomics to produce graded, copper doped beryllium shells. While these coatings have consistent microstructure and acceptable void content, we found that different coaters produced different results with respect to argon implantation. Each individual system met the requirements for argon implantation, but the deviation from one system to another and from run to run exceeded the variability requirements as specified by the NIF target design requirements. We chose a baseline coater and redesigned the fixturing within that system to enable more consistency in argon concentration from one run to the next. Then we reconfigured the coaters so that the vertical and lateral alignment of the shells under the gun varied less than 1 mm from one system to another. After this process, the systems were able to produce beryllium capsules with radial argon profiles that met specifications and were consistent from run to run and from system to system. During this process we gained valuable insight into the beryllium coating process. The radial argon variation was shown to be dependent on sputter target thickness. We also found that the argon content in the shells was extremely dependent on the *position of the shells with respect to the gun. Additionally, we have found that the absolute argon concentration can be reduced to less than 0.3 at. % with proper positioning of the capsules.*

I. INTRODUCTION

Beryllium is one of the preferred ablators for achieving ignition at the National Ignition Facility (NIF) because of its hydrodynamic stability, low opacity, thermal conductivity, and high tensile strength.[1] The fabrication of these capsules involves sputtering beryllium onto plastic mandrels, the specific coating process has been defined elsewhere.[2–4] This sputter deposition application uses argon as a vapor source; therefore, the implantation of argon is inherent to the process. Efforts to minimize the implantation of the sputtering gas are documented throughout the sputter coating community, and embedded argon levels in biased systems generally range between 1-10 at.%.[5-7] There are three sputter coating systems supporting this effort. In capsules from all of these systems, the argon concentration decreases over the course of a coating run, and spikes back up when a second run begins on the same capsules. The NIF design takes the resulting radial variation into account, and while we were able to meet the radial argon requirement in any given run, the variation from run to run was not consistent enough (Fig. 1). The average argon was 0.6 at.% and the NIF specifications permit us to vary up to 20% from the average; which allows for a range from 0.48 to 0.72 atomic percent. The observed variance was from 0.25 at.% up to 1.1 at.%. We designated one system as our baseline coater, and redesigned the fixturing to optimize our capsule and target position. During this process, we learned that the argon concentration at these levels of interest for NIF targets was critically dependent was on both the capsule position and the initial sputter target thickness. To improve our system-to-system reproducibility, we standardized our baseline process so that the coaters were identical on a sub-mm level. This required producing detailed CAD models of the coaters, which facilitated precise duplication. Over the course of this study, there were many experiments where we discovered additional sources of variability that we had not previously anticipated. After optimizing our process, we reproduced the results in our other coating systems.

II. EXPERIMENTAL

The details of the sputtering process have been described elsewhere[2-4] and only the main features are stated below. Our first set of experiments focused on producing repeatable results from one run to another within a single coating system. We designated one system which we refer to as "S2" as our baseline coater and ran a series of experiments to pinpoint the variables that influenced the argon concentration. The experiments were performed in a high vacuum chamber equipped with a 1.3 inch mini-MAC magnetron sputter gun with a beryllium target prepared from cast beryllium material.[8] In a typical experiment, the system is evacuated overnight to a base pressure of ~1×10^{-7} Torr. The argon sputtering gas is obtained from a boil off from liquid argon and scrubbed using a titanium sublimator. Sputtering is performed in a 6 mTorr argon atmosphere with the beryllium sputter gun operating at 60 W. The mandrels rotate in a tilted pan located 33 mm below the beryllium gun. The deposition system includes the ability to apply a bias voltage to the pan during the coating process. A bias of –80 V was applied. In the S2 system, the pan was screwed into a bracket that attached to the motor, which was mounted on an angled bracket. The length of the threaded rod on the pan varied and therefore the height of the pan changed by a few millimeters from one run to the next. Additionally, the motor was mounted using an oversized hole that had over a centimeter of play. We designed and fabricated a more robust set of fixtures for the chamber which corrected both of the above issues as well as some smaller sources of run to run deviation, then ran a series of experiments to quantify our reproducibility. After precisely aligning the system, we made systematic modifications to our alignment and the resulting argon concentration was measured.

II.A. Argon Concentration as a Function of Target Thickness

The mini-MAC sputter gun was fitted with a 3.8 mm thick beryllium sputter target. The GDP mandrels used were from three separate batches and had diameters of 2027 μm, 1887 μm, and 1664 μm. Mandrel diameters are measured to ±3 μm accuracy. The vertical distance from the gun to the pan was 30.4 mm, the pan tilt was measured at 12.36°, and the base pressure of the run was 2.70×10^{-7} Torr. The gun to pan distance measurement has an accuracy of ±1 mm and the angular uncertainty is ±0.2°. The pressure is accurate to 0.02×10^{-7} Torr. The initial target voltage observed for this run was 348 V. These initial target voltage measurements are accurate to ±5 V. A second run was done with the intent to observe the affect of a thicker starting material. A 4.53 mm beryllium target was installed in the sputter gun. The target thickness is measured to ± 0.08mm. GDP mandrels used were from the same batches and with the same respective diameters. The distance from the gun to the pan was once again 30.4 mm, the pan tilt was measured at 12.36°, and the base pressure of the run was 2.83×10^{-7} Torr. The initial target voltage observed for this run was 381 V.

II.B. Argon concentration as a Function of Diameter

The investigation of the affect of mandrel size on argon implantation took place in many runs. A number of runs were performed with a wide variety of mandrels of various diameters inserted into the pan. The three sizes of mandrels we focused on were chosen because of their relevance to the baseline design for the NIF beryllium target. The diameters of interest were

1600 μm, 1800 μm, and 2000 μm. The mandrel diameters were documented for each run and identified afterwards based on their respectively different sizes. Subsequent characterization of capsules with respect to individual diameters was performed. These tests were performed in all three coaters under a variety of configurations and the effects were reproducible.

II.C. Argon concentration as a Function of Darkshield Positioning

The sputter coating gun itself has a darkshield, which serves to protect the magnets and the ceramics in the gun from arcing damage. It is essentially a ceramic sheath that isolates the gun's electric field. It is located in the dark space, an area of high voltage potential which surrounds the target. The darkshield was initially installed by hand with the vertical spacing determined by feel. Three fixtures were fabricated to accurately set the height of the darkshield with respect to the targets. The darkshield to target height was set at 1.2 mm, 2.5 mm, and 3.0 mm with a measurement uncertainty of ±0.06 mm. The runs were performed using 3.8 mm thick beryllium targets, and the base pressure was below 4.5×10^{-7} for all of the runs. The distance from the gun to the pan was 30.4 mm, the pan tilt was measured at 12.36°. The initial target voltage observed in these runs did not vary significantly; it ranged from 366 V with 3.0 mm darkshield spacing to 358 V when the darkshield was spaced at 1.2 mm.

II.D. The Effect of Mandrel Quantity on Argon Concentration

We investigated the effect of mandrel quantity on the argon implantation in multiple systems, as we did with the mandrel diameter study. A number of runs were performed with either 15 or 6 shells in the pan in the various coating systems. Subsequent characterization of the capsules with respect to the quantity in the pan was performed. The initial target voltage observed did not change significantly with changes in mandrel quantity.

II.E. Argon Concentration as a Function of the Distance Between the Gun and the Pan

We performed an experiment where we decreased the distance between the beryllium gun and the coating pan. We decreased the gun to pan distance from 30.4 mm to 27.3 mm; and then again to 26.9mm. Due to the configuration of the system, it would be extremely difficult to decrease the distance further. We set the darkshield height at 2.5 mm, the pan angle was held constant at 12.36°, and the base pressure was below 4.5×10^{-7} for all of the runs. The initial target voltages ranged from 365 to 372 Volts for these runs.

III. RESULTS

The characterization plan for these experiments included verification that the modifications did not result in unanticipated changes to the capsule in areas where we already met specifications. Following the standard plan for NIF capsule metrology,[9] we characterized shell wall thickness, outer diameter, density, and wall thickness uniformity. The shells were measured for out-of-roundness using an optical microscope and the RMS surface finish was determined using a "spheremapper," which is a modified AFM that acquires equatorial traces in order to produce a power spectrum.[10] The chemical composition of argon was measured using quantitative X-Ray radiography, X-Ray fluorescence, Energy-dispersive X-ray spectroscopy (EDS) and edge spectroscopy. All of these methods have been validated for this process.[11-14] The X-ray radiography can measure to an accuracy of 0.05 at.%. X-Ray fluorescence is accurate to within 20% and gives a bulk concentration. EDS has an error bar of ±0.0.1 at.%, and edge spectroscopy give a bulk dopant value that has an error bar of 0.1 at.%. When used in conjunction with each other, these techniques give an overall error of ±0.05 at.% Ar.[14] Our report focuses on the argon concentration in the shells but we characterized the other parameters in order to watch for unforeseen consequences.

The coating conditions in all three coaters were on occasion radically changed and the results of those runs were very different from the conditions documented above; those results are not included here. In general, coating runs followed a pattern where the voltage and the argon concentration decreased over the course of the run. The general trend of both the argon and the voltage is illustrated in Fig. 2. We attribute this to the fact that as the target erodes, the plasma gets closer to the magnets where the magnetic field is stronger. The increased electron confinement leads to higher density plasma. The resultant increase in the plasma conductivity leads to a lower voltage and higher current for the same input power to the gun. But due to the lower accelerating voltage, the argon ions impinging on the capsule are not as energetic leading to a decrease in the argon concentration.[6]

III.A. Argon Concentration as a Function of Target Thickness

The results of the target thickness experiment are documented in Fig. 3. Note the shape of the radial argon concentration profile. The argon concentration begins to descend from its highest levels after about 10 μm of coating; between 20 μm and 30 μm it descends below 0.2 at.%. The shape of this profile is characteristic of all our experiments. In this experiment, the initial argon concentration of the shells coated with the 4.5 mm target is more than 0.2 at. % greater than that of the shells coated with the 3.8 mm target. The thinner target resulted in lower argon values, both initially and as the coating progressed. This is consistent with the theory reported above.

With the increased target thickness, the plasma starts out even farther from the magnets and is less dense. This results in higher voltage, increased argon energy, and more implantation.[6, 7]

III.B. Argon Concentration as a Function of Diameter

The concentration of argon in the shell as related to various mandrel sizes is illustrated in Fig. 4. The trend was seen in many different runs and in all three coating systems. The initial argon concentration was ~0.4 at. % higher in the ~1600 μm mandrels as compared to the ~2000 μm mandrels. Over the course of the coating run, the argon value for all three mandrel sizes converged, and at 25 μm coating thickness the difference in argon concentration between the shells was less than 0.1 at. %. At this time we do not have a clear understanding as to what mechanism causes this difference.

III.C. Argon Concentration as a Function of Darkshield Positioning

The darkshield was tested in three different positions and with three mandrel sizes, ranging from ~1600 μm to ~2000 μm. In Fig. 5, the first graph shows the initial argon concentration with respect to both the mandrel size and the darkshield position. As anticipated from our results described in III.B, the smallest mandrels had the greatest initial argon concentration. The second graph shows the final argon values and their relation to the mandrel size and the darkshield position. We did not observe a relationship between the final argon concentration and darkshield position or mandrel diameter. As the target is used up during the coating process the plasma gets closer to the magnets and appears to be less influenced by the position of the darkshield. The shape of the magnetic field in the sputtering process is complex and while we assume that the darkshield influences the field and therefore the argon energies, we do not have a model to explain this effect.

III.D. The Effect of Mandrel Quantity on Argon Concentration

All of our initial test runs were done with six capsules with two of each mandrel size. We decided to observe the changes that would result when we increased the number of mandrels in the coater from six to fifteen. Fig. 6 shows the results of this experiment. The initial argon concentration and the average argon concentration were always higher when there were more mandrels in the pan. These experiments were done in all three coating systems before variability was reduced with reliable fixturing, and therefore there is a wider range of results. Increasing the number of mandrels can lead to one obvious change. During coating, the mandrels stay as a group on the rotating pan and randomly share the center and the peripheral positions. As the number of mandrels increases, so does the area in the pan where the shells reside. Thus, mandrels spend more time in positions where the angle of the deposition is non-optimal, leading to higher argon.

III.E. Argon Concentration as a Function of the Distance Between the Gun and the Pan

The vertical distance between the gun and the pan was decreased by 3.1 mm to study the change in argon concentration. Fig. 7 shows that when the gun was closer to the pan, the argon concentration started out at a lower value. Conversely, as the coating thickness increased, the argon concentration was higher when the gun was closer to the pan. The voltage for both runs is also plotted, and is only slightly different between these two runs. This indicates that the plasmas had similar densities. We believe that early in the run, when the target is in close proximity to the pan, the mandrels are so close to the plasma that there is very little room for the argon ions to approach the substrate. As the target erodes, the plasma is located farther away from the pan. At that point, the mechanism for argon implantation is more dominated by the number of collisions the argon atoms undergo as well as their respective energies. The voltages of these two runs were similar, which indicates the dominating factor was the number of collisions. As the pan distance is decreased, the argon ions lose less energy through collisions as they travel through the plasma to deposit on to the capsule. Since argon incorporation into the beryllium coating should be proportional to the argon ion energy[6], the average argon concentration is greater when the gun is closer to the pan.

This would imply that moving the pan even closer to the gun could result in a coating where the argon concentration was limited by volume. We performed a set of runs with the gun to pan distance at 26.9mm. The results are shown in Fig. 8. The argon concentration started at less than 0.3 at% and remained low throughout the coating run. Decreased argon contamination is desirable, but there are other variables (roundness, leak rate, surface finish) to consider. If these shells meet specifications in other regimes, we may again reconfigure our coaters to match these conditions and reproduce these results. To accommodate this change, we need to modify many of the brackets and assemblies in the other coaters as they were not designed to support this setup. We are in the process of fully characterizing these capsules to determine if it would be desirable to implement this change on all three coaters.

III.F. Integration of Results into Other Systems

Compiling our results led to conclusions which would allow us to tune the coaters to maintain more consistent argon results. We considered the results and their implications with respect to production throughput and design specifications.[15] Thinner targets could result in lower argon values. Based on our current coating configuration and a 3.8 mm thick target, a full thickness capsule will take four separate coating runs with each run lasting 4–5 days. While a thinner target could lead to lower initial and total argon values, the resulting production flow would be impractical. It was determined that the 3.8 mm target provided enough material while still remaining marginally practical for production, but that a thinner target would not be adequate in terms of production rates. We demonstrated that the argon concentration can be controlled by the mandrel diameter, but the size of the mandrel is determined by the final beryllium capsule specifications and cannot be controlled except to modify the final target design. While we cannot modify this parameter, experiments showed that the trend was repeatable and if the results are consistent they can meet the design specifications. Prior to understanding the effect of mandrel size on the argon concentration, we were having difficulty understanding our experimental results. Using the knowledge we gained from the darkshield positioning experiments, we standardized our process by setting the darkshield at 3.1 mm in every run in order to minimize the initial argon. In order to efficiently provide capsules for NIF laser experiments, reducing the mandrel quantity to less than 15 shells per run would significantly decrease our productivity. We again used this information to interpret other results, but did not modify our coating plan to decrease the number of mandrels in a production coating run. Finally, we chose a gun-to-pan distance that could be accommodated in all three coating systems. Previous work has made us wary of increasing the proximity of the shells to the target because the mandrels can deform in these high temperature situations. We made detailed CAD designs of all three coating systems and developed fixtures, jigs, and hard stops that assisted with repeatable alignment within the chambers. The results of this work are shown in Fig. 9, which plots final data for a number of runs resulting in full thickness capsules in two of the three coating systems.

IV. CONCLUSION

By performing a series of experiments using carefully measured modifications, we have been able to optimize the coater configuration in such a way as to maintain consistent, repeatable argon profiles that meet the NIF design specifications for beryllium targets.[14] We minimized the initial argon value by modifying the gun-to-pan distance and the darkshield position. We also determined an optimal thickness for our initial target and the quantity of mandrels that can be successfully coated at one time. Using this data, we have been able to reproduce these results in the three systems that are dedicated to this effort. These shells meet the NIF specifications and are reproducible in all three coating chambers.

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FIGURES AND CAPTIONS

Fig. 1. Atomic Percent Argon Vs. Coting Thickness. The NIF spec is indicated by the shaded band. NIF specifications were not consistently met from run to run in the coating systems.

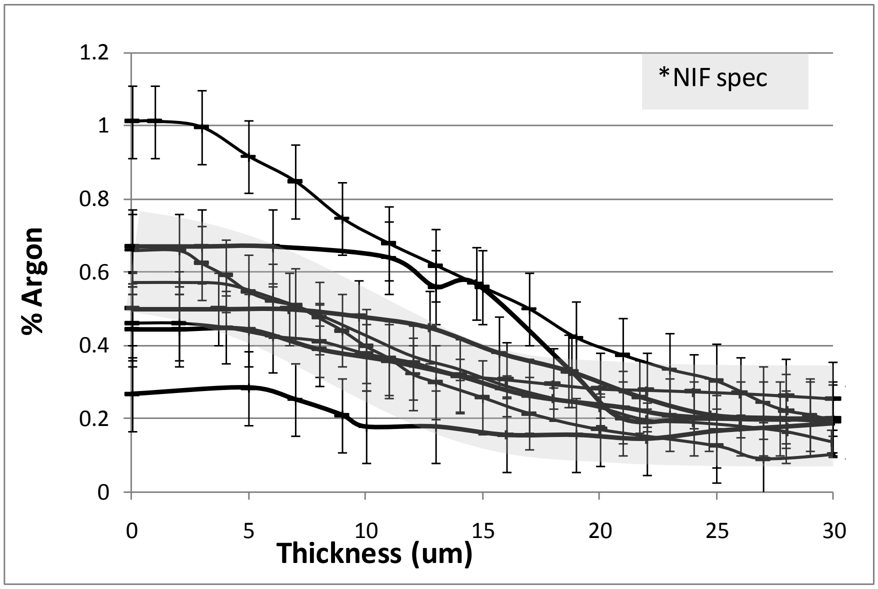

Fig. 2. Typical Voltage and Argon at. % curves plotted with respect to coating thickness.

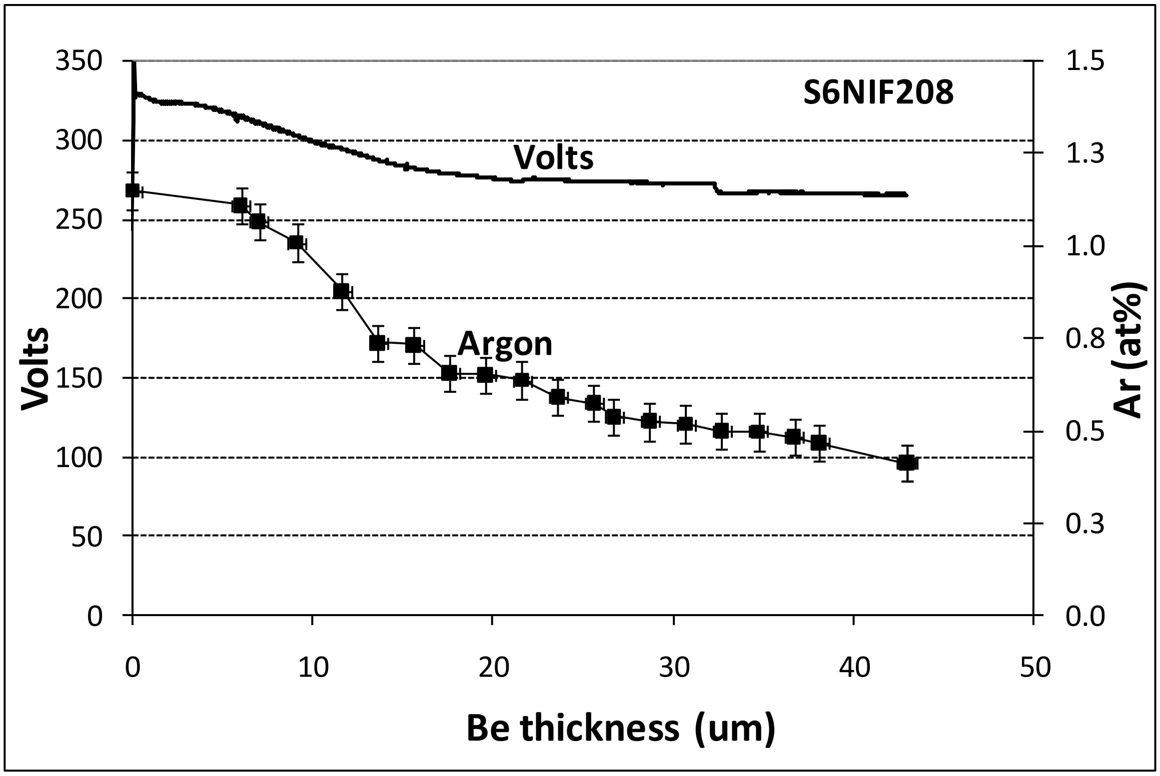

Fig. 3. A typical curve showing argon concentration as a function of target. Thicker targets result in higher argon.

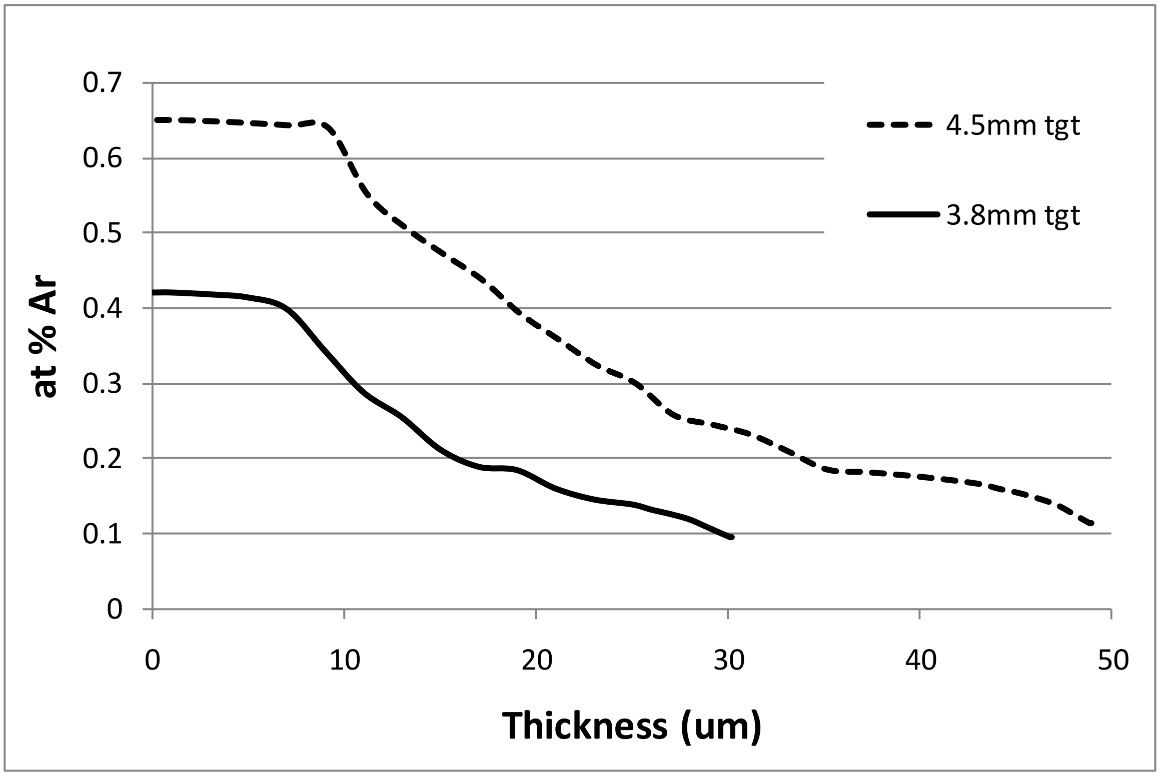

Fig. 4. Argon as a f(x) of mandrel diameter.

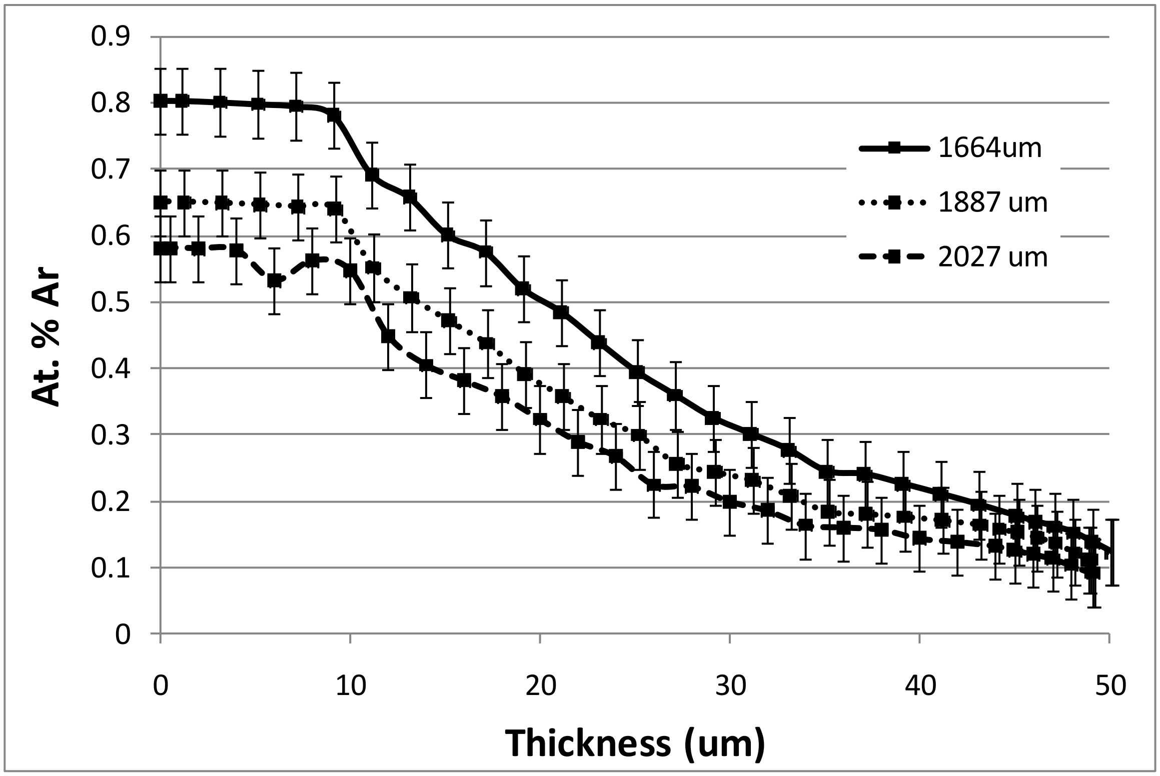

Fig. 5. Initial and final argon as a f(x) of DS position. The graph on the left shows the initial argon values of different mandrel sizes organized by the darkshield spacing. The plot on the right shots the argon concentration at the end of a coating run of the various mandrel sizes; again organized by the darkshield spacing. While there is a trend that can be seen in the first graph, the darkshield jig does not have an apparent effect on the final argon concentration.

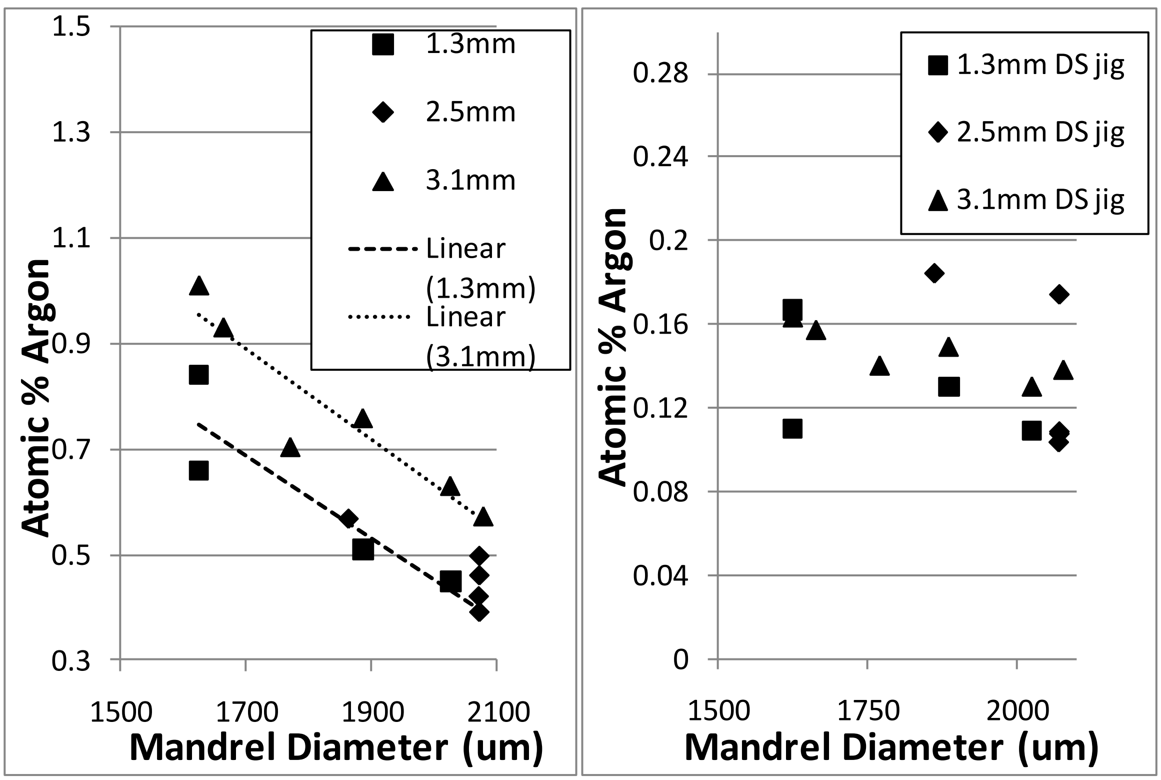

Fig. 6. The plot on the left shows the initial argon concentration as a function of shell quantity; and the plot on the right shows the average argon concentration as a function of shell quantity. The graphs illustrate that both the initial and average argon are elevated when more shells are in the coating pan.

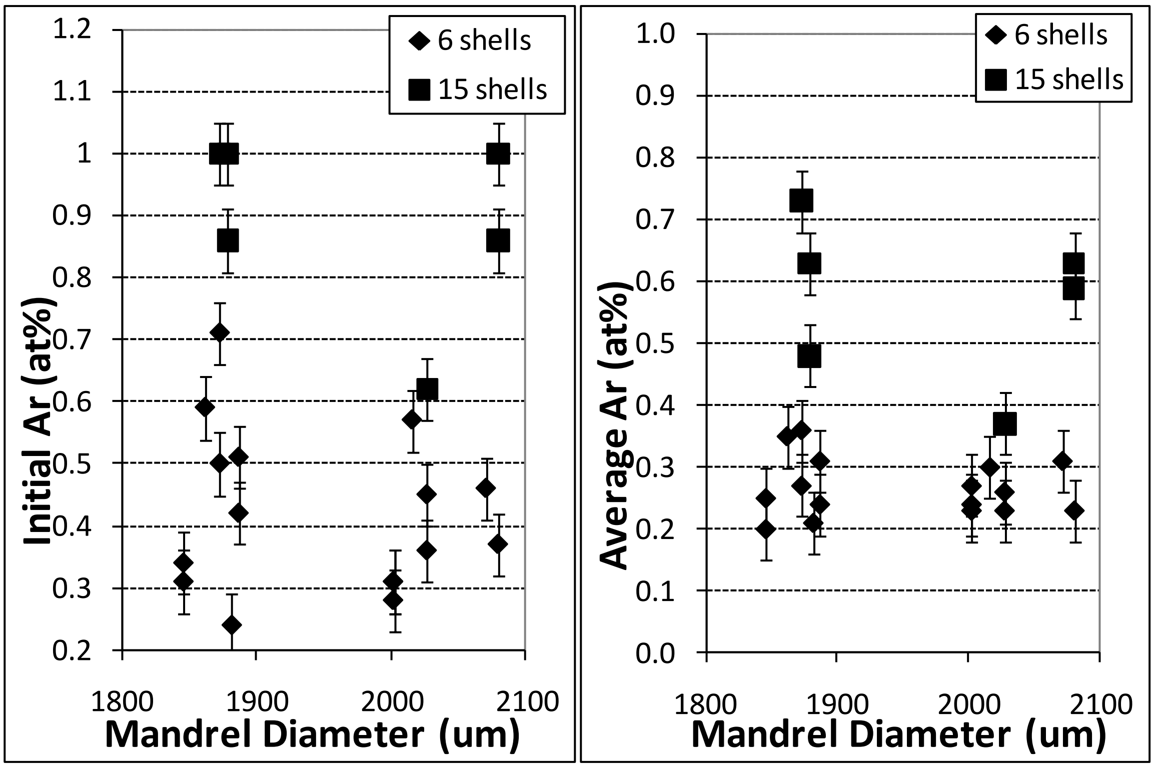

Fig. 7. Argon concentration as related to the distance from gun to pan.

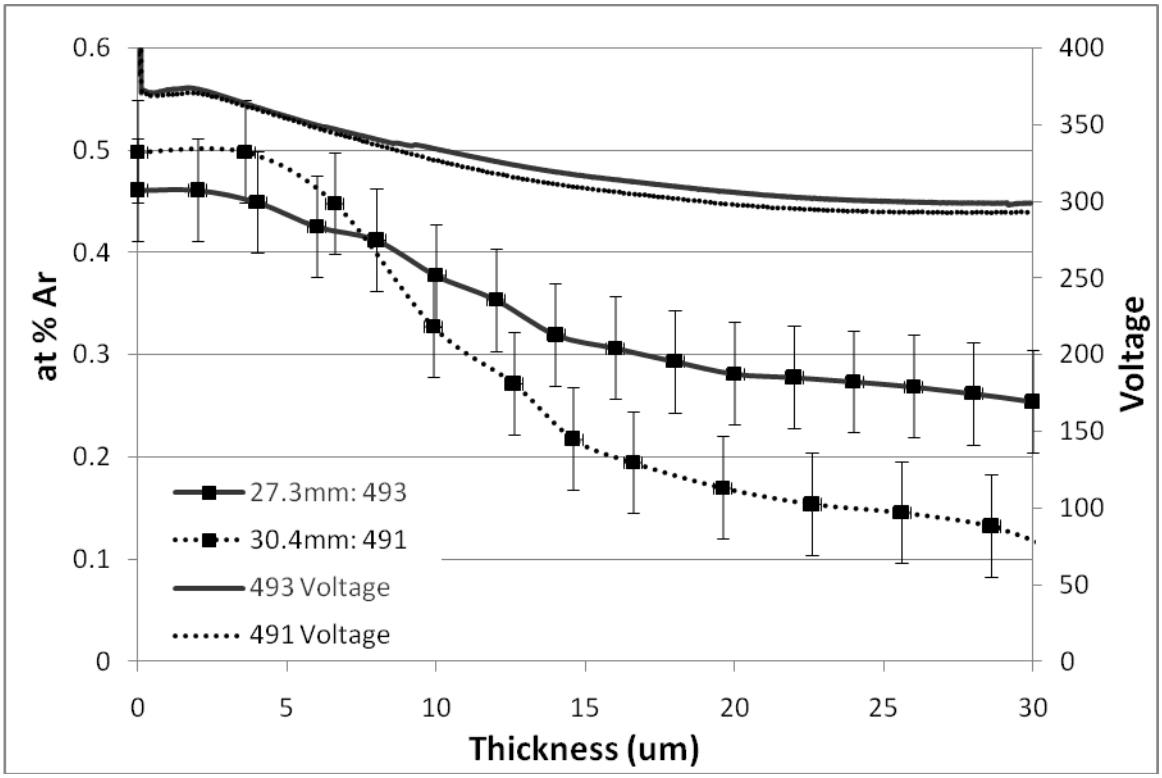

Fig. 8. Argon profiles in coater with 26.9 mm gun-to-pan distance resulted in ~35% lower initial argon levels.

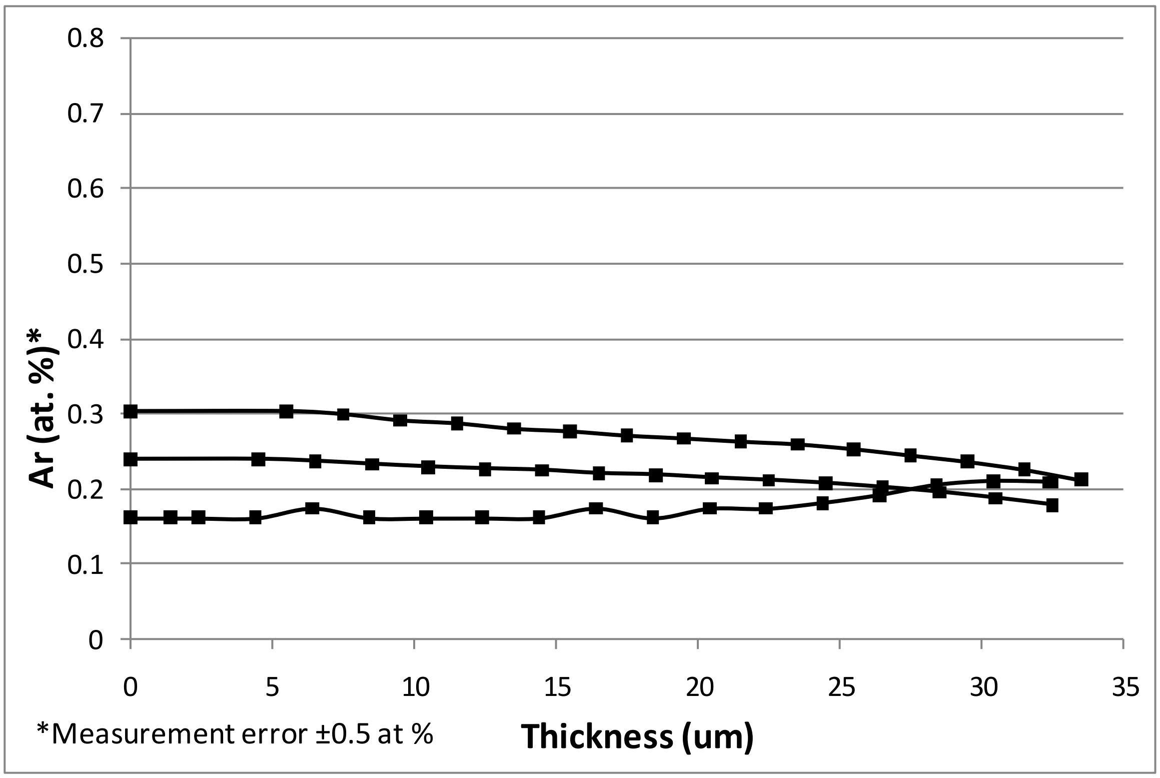

Fig. 9. Results of optimization. It should be noted that the first 5um of coating are difficult to measure due to surface interference.